Torque magnetometry of YBCO crystals above T_c

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The magnetization of three small high-quality single crystals: $YBa_2Cu_3O_7$, $YBa_2Cu_3O_{6.5}$ and $YBa_2Cu_3O_{6.36}$ has been measured using torque magnetometry. Most of the data are consistent with the theory of Gaussian superconducting fluctuations, together with a strong cut-off above $T \gtrsim 1.1T_c$. The effect of inelastic scattering could be an important ingredient for achieving detailed understanding.

Cuprate superconductors show much stronger thermodynamic fluctuations than classical ones because of their higher transition temperatures (T_c), shorter Ginzburg-Landau (GL) coherence lengths and quasi-two dimensional (2D) layered structures with weakly interacting CuO_2 planes [1, 2]. Observations of diamagnetism [3] and large Nernst coefficients over a broad temperature (T) range well above T_c for several types of cuprate [4, 5] are intriguing [6] and could provide evidence for unusual vortex-like superconducting excitations. On the other hand calculations [7] using the lowest-order quadratic or Gaussian fluctuation (GF) term in GL theory, account for Nernst data for optimally (OP) and over-doped (OD) $La_{2-x}Sr_xCuO_4$ (LSCO) crystals. While for an under-doped (UD) crystal, they suggest that critical fluctuations reduce T_c from its mean field value, T_c^{MF}, and give a larger Nernst signal than GF near T_c. Later, it was argued that Nernst data for Ca and Zn-doped $YBa_2Cu_3O_{6+x}$ (YBCO) [8] and the published data for LSCO [4], are mostly consistent with GF theory [7].

One difficulty in this area is separating the fluctuation (FL) contribution to a given property from the normal state (N) background. Recently this has been dealt with for the in-plane electrical conductivity $\sigma_{ab}(T)$ of YBCO crystals by applying very high magnetic fields (B) [9]. When analyzed using GF theory, $\sigma_{ab}^{FL}(T)$ was found to cut off even more rapidly above $\epsilon \equiv (T - T_c)/T_c = 0.1$ than previously thought [10, 11]. It was also strongly reduced at high B and the fields needed to suppress $\sigma_{ab}^{FL}(T)$ extrapolated to zero between 120 and 140 K depending on x, which tends to support a vortex or Kosterlitz-Thouless scenario. Another related puzzle is the strong and unexpected symmetry of the heat-capacity anomalies for all UD cuprates. Recently this has been explained [12] in terms of a large difference between T_c^{MF} and T_c, for example T_c^{MF} = 90 K for Ca-doped UD60 YBCO, where in the notation used here the low-field T_c = 60 K. Therefore questions such as the applicability of GF theory $vs.$ a phase fluctuation (vortex) scenario and the extent to which T_c is suppressed below T_c^{MF} by strong critical fluctuations, are still being discussed.

Here we report torque magnetometry data measured [13] from T_c to 300 K for tiny YBCO single crystals from OD to heavily UD, grown in non-reactive $BaZrO_3$ crucibles from high-purity (5N) starting materials. We analyze the results using GF theory which, unlike some other approaches, predicts the *magnitude* of the observed effects as well as their T-dependence. Although measurements of the London penetration depth [14] below T_c and thermal expansion [15] above and below T_c for OP YBCO crystals, give evidence for critical fluctuations described by the 3D-XY model, up to ± 10 K from T_c, we argue later that these do not alter our overall picture.

When B is applied parallel to the c-axis the GF contribution to the magnetization along c is [2]:

$$M_c^{FL}(T) = -\frac{\pi k_B T B}{3\Phi_0^2} \frac{\xi_{ab}^2}{s\sqrt{1 + (2\xi_{ab}/\gamma s)^2}} \qquad (1)$$

Here $\gamma = \xi_{ab}/\xi_c$ is the anisotropy, $\xi_{ab} = \xi_{ab}(0)/\epsilon^{1/2}$ and $\xi_c = \xi_c(0)/\epsilon^{1/2}$ are the T-dependent GL coherence lengths $\parallel$ and $\perp$ to the layers, s = 1.17 nm is the distance between CuO_2 bi-layers, Φ_0 is the flux quantum for pairs and k_B is Boltzmann's constant. Eq. 1 is valid when $B \ll \Phi_0/(2\pi\xi_{ab}^2)$ and then the susceptibility $\chi_c^{FL} \equiv M_c^{FL}/B$ does not vary with B. For $B \perp c$, $\chi_{ab}^{FL} = 0$ in the 2D limit $s \gg \xi_c(T)$, and in the opposite 3D limit $\chi_{ab}^{FL}(T) = \chi_c^{FL}(T)/\gamma^2$ [1]. The torque density $\tau \equiv \underline{M} \times \underline{B}$ and if $M \propto B$, $\tau(\theta) = \frac{1}{2}\chi_D(T)B^2 \sin 2\theta$, where θ is the angle between B and the CuO_2 planes and $\chi_D(T) \equiv \chi_c(T) - \chi_{ab}(T)$.

Much of our data, including the two curves for UD57 in Fig. 1 at higher T do obey $\tau(\theta) \propto B^2 \sin 2\theta$. However as shown in Fig. 1, there are striking deviations at lower T which are discussed later. Fig. 2a shows $\chi_D(T)$ obtained from $\sin 2\theta$ fits to $\tau(\theta)$ for 3 doping levels at high enough T so that $M \propto B$. The dashed lines show the normal state background anisotropy $\chi_D^N(T)$ which arises from the g-factor anisotropy of the Pauli paramagnetism [16]. For UD crystals the T-dependence of $\chi_D^N(T)$ is caused by the pseudogap (PG) [17] plus a smaller contribution from the electron pocket [16] observed in high field quantum oscillation studies [18]. OD89 has no PG and presumably no pockets, so we represent $\chi_D^N(T)$ by the second order polynomial in T shown in Fig. 2a.

The solid lines in Fig. 2a show fits of the $\chi_D(T)$ data for OD89 and UD57, to $\chi_D^N(T)$ plus $\chi_c^{FL}(T)$ given by Eq. 1 but with $\epsilon = \ln(T/T_c^{MF})$, as is usually done for GF fits over a wide range of T [2, 9]. We only need to consider χ_c^{FL} because the contribution from χ_{ab}^{FL} is negligible even for OD89 where $\gamma = 5$ [19]. However $\chi_c^{FL}(T)$ must be cut off for $T \gg T_c$ and therefore we multiply Eq. 1 by the same exponential attenuation factors found for $\sigma_{ab}^{FL}(T, B)$ [9] that

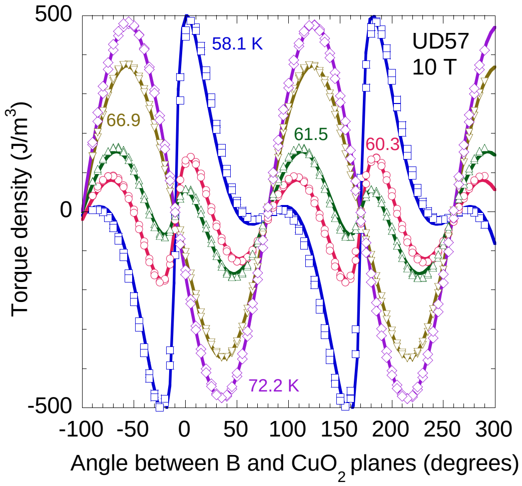

FIG. 1: Color online. Angular dependence of the torque density for the UD57 $YBa_2Cu_3O_{6.5}$ crystal in 10 T at T= 58.1, 60.3, 61.5, 66.9 and 72.2 K. The solid lines show single parameter fits to the formula for 2D GF derived from Eq. 2. Note the $\sin 2(\theta+\alpha)$ behavior at higher T, where α =10° is a fixed instrumental offset.

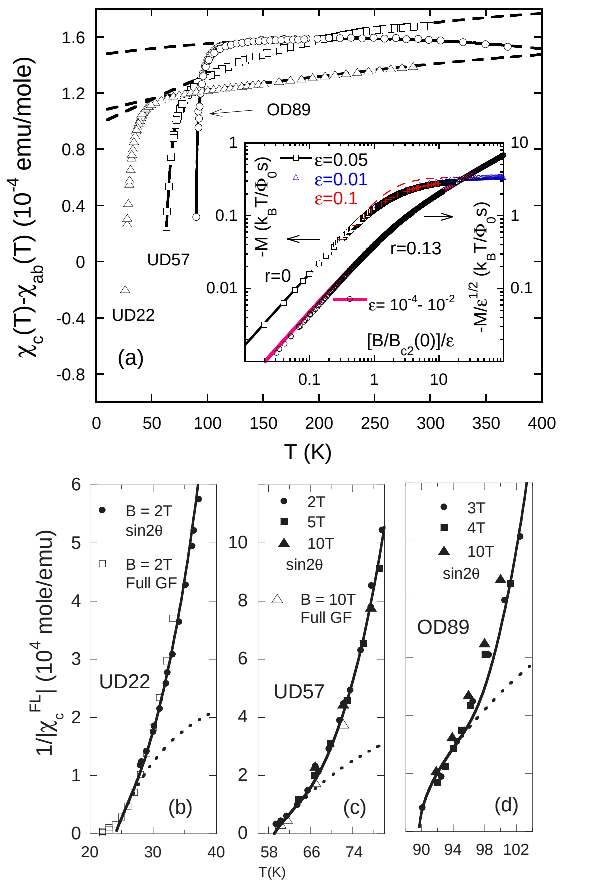

FIG. 2: Color online: (a) Main: $\chi_D(T)$ for the three crystals, dashed lines show $\chi_D^N(T)$. Solid lines for OD89 and UD57 include $\chi_c^{FL}(T)$ given by Eq. 1 and a strong cut-off [20].
Insert: Symbols show M calculated for various values of ϵ, using Eq. 2, when the anisotropy parameter $r \equiv (2\xi_c(0)/s)^2$ = 0. For r= 0.13 symbols show M given by the 2D-3D form of Eq. 2, which contains r and an extra integral [2]. The lines show formulae used [21] to represent these values of M when fitting $\tau(\theta)$ data.
(b) to (d) - plots of $1/|\chi_c^{FL}|$ vs. T for the three crystals. GF fits based on Eq. 1, are shown by short dashed lines, without a cut-off and by solid lines, with a strong cut-off [20]. Open squares for UD22 and open triangles for UD57, show $\xi_{ab}(0)^2/\epsilon$ (converted to $1/|\chi_c^{FL}|$ using Eq. 1) obtained by fitting $\tau(\theta)$ to the full 2D GF formula when $M(B)$ is non-linear.

come in above $\epsilon \simeq 0.1$. These are listed in footnote 20. For UD22 there are no $\sigma_{ab}^{FL}(T,B)$ data so Fig. 2a shows only $\chi_D^N(T)$. Figs. 2b to 2d show plots of $1/|\chi_c^{FL}(T)|$ vs. T where $\chi_c^{FL}(T) \equiv \chi_D(T) - \chi_D^N(T)$. Within GF theory these plots show where $\chi_c^{FL}(T)$ diverges and thereby define T_c^{MF}, the temperature at which the coefficient of the $|\psi|^2$ term in the GL free energy changes sign. The short-dashed lines for UD22 and UD57 in Figs. 2b and 2c show the contribution from Eq. 1 in the 2D limit ($\gamma \to \infty$) with the single adjustable parameter $\xi_{ab}(0)$ = 4.5 and 2.02 nm respectively, while the solid lines show the effect of the cut-off [20]. For OD89 we use Eq. 1 with $\xi_{ab}(0)$ = 1.06 nm and γ = 5, [19] shown by the short-dashed line, with the solid line again including the cut-off [20]. The high quality of these fits could be somewhat fortuitous in view of our neglect of any charge density wave (CDW) [17], but for UD57 other subtraction procedures give similar values of $1/|\chi_c^{FL}(T)|$. Parameters obtained in the present work are summarized in Table I. Heat capacity studies give a very similar value $\xi_{ab}(0)$ = 1.12 nm for OD88 YBCO [22] while our values for UD57 and UD22 agree with previous work [9, 23] for the same T_c values. For UD57, setting $\gamma = 45$ [24], rather than the 2D limit of Eq. 1 ($\gamma \to \infty$) has no significant effect.

As the critical region is approached from above T_c the exponent of $\xi_{ab}(T)$ is expected to change from the MF value of -1/2 to the 3D-XY value of -2/3 [1]. It is very likely that this will also apply to strongly 2D materials, including UD57, since heat capacity data above and below T_c [25] do show the $\ln|\epsilon|$ terms associated with the 3D-XY model. We have addressed this by repeating our GF fits in Figs. 2b and 2c with $\epsilon \geq 0.20$ (UD22) or 0.15 (UD57) without altering the cut-off [20]. The only significant change is that $\xi_{ab}(0)$ becomes 15% larger for UD57. For OD89 the width of the critical region is much smaller than for OP YBCO [14, 15] because of the extra 3D coupling arising from the highly conducting CuO chains [22]. Fits to GF theory for OD89 with T_c^{MF} = 90 K and $\epsilon \geq 0.05$ do not alter $\xi_{ab}(0)$ within the quoted error.

Fig. 3 shows plots of $\tau/B\cos\theta$ vs $B\sin\theta$ at fixed T for UD57. We use this representation of the data and MKS units, A/m, for comparison with Ref. 3. If $\chi_D^N(T)$ is subtracted, which has not been done for Fig. 3, then for 2D fluctuations this would be the same as plotting M_c^{FL} vs. $B \parallel c$. Near T_c there is substantial B dependence which as shown in detail below is consistent with GF theory. However GF formulae are expected to be approximately valid even in the crossover region to 3D-XY behavior [1], to first order the main effect is

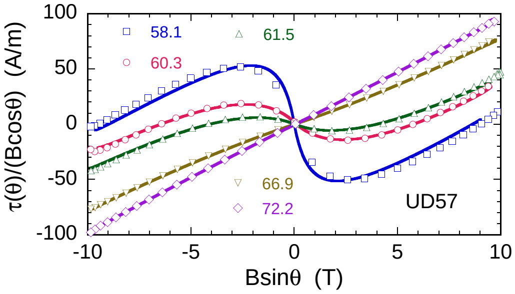

FIG. 3: Color online: Magnetic field dependence of the magnetization obtained from the torque data for UD57 at T = 58.1, 60.3, 61.5, 66.9 and 72.2 K. Solid lines show fits to the 2D GF formula for M plus the same normal state contribution used in Figs. 1, 2a and 2c.

the change in the exponent of $\xi_{ab}(T)$. For GF in the 2D limit, the free energy density at all B is [2]:

$$F = \frac{k_B T}{2\pi \xi_{ab}^2 s}\{b\ln[\Gamma(\frac{1}{2}+\frac{\epsilon}{2b})/\sqrt{2\pi}]+\frac{\epsilon}{2}\ln(b)\} \quad (2)$$

using the standard Γ function, with $b = B/\tilde{B}_{c2}(0)$, where $\tilde{B}_{c2}(0) = \Phi_0/2\pi\xi_{ab}(0)^2$, and as before $\epsilon = [T - T_c(B = 0)]/T_c(B = 0)$. The magnetization $M = -\partial F/\partial B$ obtained by numerical differentiation of Eq. 2 for three typical values of ϵ is shown in the insert to Fig. 2a. It can be seen that M, in units of $k_BT/\Phi_0 s$, scales with b/ϵ to within a few % and can be adequately represented by the simple formula $-b/(3b+6\epsilon)$, that has a single unknown parameter $\xi_{ab}(0)^2/\epsilon$. Figs. 1 and 3 show it gives a good fit to our data for UD57 and, importantly, when these values of $\xi_{ab}(0)^2/\epsilon$ are inserted into Eq. 1, then as shown by the open triangles in Fig. 2c, they give $1/\chi_c^{FL}(T)$ values that agree well with points from $\sin 2\theta$ fits at lower B or higher T. For OD89 strong deviations from $\sin 2\theta$ behavior only occur within ~ 1 K of T_c and these [26] are not properly described by GF theory. For UD22, $\tau(\theta)$ plots in 2T at low T (not shown) also give $\xi_{ab}(0)^2/\epsilon$ values that overlap nicely with the high T data as shown in Fig. 2b. However small jumps in $\tau(\theta)$ at $\theta = 0$ between 33 and 26 K of size $M_c = 0.01 - 0.02 k_BT/\Phi_0 s$ could not be fitted. They are ascribed to regions with higher T_c [27], 1-2% of the total volume, that are much smaller than the London penetration depth, so they give negligible screening at low fields.

The good description of our data by this GF analysis suggests that the high critical fields proposed in Refs. 3–5 for $0.01 < \epsilon \lesssim 0.2$ are <u>not</u> fields needed to suppress diamagnetism associated with vortex-like excitations. Instead these fields show where the positive linear term in $\tau/B\cos\theta$ from χ_D^N compensates the field-independent high-field diamagnetism from 2D GF, $M \simeq -0.33k_BT/\Phi_0 s = -0.112$ emu/cm^3 or -112 A/m at 60 K. They are several tens of Tesla depending on s, γ, the GF cut-off and $\chi_D^N(T)$, which in turn depends on the size of the PG [16]. We note that the present results are consistent with a recent study of B_{c2} for YBCO [28] and that recent torque magnetometry data [29] for various single layer cuprates, including Hg1201 show similar exponential attenuation factors to those for YBCO [9, 20].

An intriguing question about the present results and those of Ref. 9 is the origin of the strong cut-off in the GF above $\sim 1.1T_c$. If the weakly T-dependent $\chi_D^N(T)$ behavior for OD89 shown in Fig. 2a is correct then both our $\chi_D^{FL}(T)$ data and $\sigma_{ab}^{FL}(T)$ [9] decay as $\exp[-(T-1.08T_c)/T_0]$ above $T \sim 1.08T_c$ with T_0 ~9 K. If instead $\chi_D^N(T)$ were actually constant below 200 K then our $\chi_D^{FL}(T)$data would give a slower decay than Ref. 9, namely T_0 ~25 K. In either case the presence of this cut-off for OD YBCO samples seems to rule out explanations connected with the mean distance between carriers since in the absence of a PG the hole concentration is expected be $\simeq 1.2$ per CuO_2 unit, as shown directly by quantum oscillation studies of OD $Tl_2Ba_2CuO_{6+x}$ crystals [30].

Assuming there are no unsuspected effects caused by d-wave pairing, one hypothesis is that the GF and possibly T_c itself are suppressed by inelastic scattering processes. In a quasi-2D Fermi liquid the T-dependence of the electrical resistivity is determined by the circumference of the Fermi surface and the inelastic mean free path, l_{in}. For OD YBCO the measured a-axis resistivity [23] gives l_{in} = $2.5(100/T)$ nm, but values for UD samples are less certain because of the PG. The BCS relation $\xi_{ab}(0) = \hbar v_F/\pi\Delta(0)$, where $\Delta(0)$ is the superconducting energy gap at $T = 0$, implies that irrespective of the value of the Fermi velocity v_F, the usual pair-breaking condition for significant inelastic scattering, $\hbar/\tau_{in} \gtrsim \Delta(0)$ is equivalent to $l_{in} \lesssim \pi\xi_{ab}(0)$. This is satisfied at 100 K for OD YBCO. So some suppression of GF and indeed T_c by inelastic scattering does seem to be entirely plausible. If T_c is suppressed then $\Delta(T)$ will fall more quickly than BCS theory as T_c is approached from below, which would affect the analysis of Ref. 12.

Another possibility [9] which might account for the observations, is that the pairing strength itself falls sharply outside the GL region, for example when the in-plane coherence length becomes comparable to, or less than, the correlation length of spin fluctuations. From Figs. 2b to 2d we can read off the values of T where the solid and dashed lines differ by (say) a factor of two. At these points $\xi_{ab}(T) \equiv \xi_{ab}(0)/\ln(T/T_c^{MF})$ = 11.9, 9.5 and 7.9 nm for UD22, UD57 and OD89 respectively. Neutron scattering studies [31, 32] typically give a full width half maximum of $0.17\frac{2\pi}{a}$ for the scattering intensity from spin fluctuations. Although this does vary with composition and scattering energy it corresponds to a correlation length [33] of just over 6 lattice constants, a, or 2.5 nm, similar to $\xi_{ab}(0)$ but much smaller than the $\xi_{ab}(T)$ values for which χ_c^{FL} is reduced by a factor two. It remains to be seen whether theory could account for this.

In these pictures the effective T_c describing the strength of the GF would fall for $T > 1.1T_c$ either because of inelastic scattering or because of a weakening of the pairing interaction. If it could be shown theoretically that $\tilde{B}_{c2}(0)$ falls in a similar manner, this would account naturally for the fact [9] that the magnetic fields needed to destroy the GF fall to zero in the temperature range 120-140 K, where the fluctuations become very small, and would be decisive evidence in favor

$Sample$	$^{\S}T_c$ (K)	T_c^{MF} (K)	$\xi_{ab}(0)$ (nm)	$0.59\tilde{B}_{c2}(0)^{\ddagger}$ (T)	$\Delta(0)^{\dagger}$ (K)
OD89	89.4	89.7	1.06 ± 0.1	173	448
UD57	56.5	59	2.02 ± 0.1	48	234
UD22	21.6	24	4.5 ± 0.5	10	105

TABLE I: Summary of results. $^{\S}T_c$ defined by sharp onsets of SQUID signal at 10G and torque data at $\pm$50G. ‡2D clean limit formula [2] for $B_{c2}(0)$. †From the BCS relation $\xi_{ab}(0) = \frac{\hbar v_F}{\pi\Delta(0)}$, which may not hold exactly for d-wave pairing, with v_F=2x10^7 cm/sec.

of the GF scenario.

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[21] The solid line for $r = 0$ shows our empirical 2D formula $b/(3b + 6\epsilon)$, where $b = 2\pi\xi_{ab}(0)^2B/\Phi_0$. The dashed line shows the 2D limit of Eq. 1 with $\xi_{eff}(b)$ given by $\xi_{eff}(b)^{-4} = \xi_{ab}(T)^{-4} + l_B^{-4}$, where $l_B = (\hbar/eB)^{1/2}$, the formula used to analyze Nernst data for NbSi films [36]. For $r = 0.13$, $b < r$ and $\epsilon < r$, our empirical 3D formula is $-M/\sqrt{\epsilon} = (k_BT/s\Phi_0)0.68b/\sqrt{\epsilon(b+1.94\epsilon)}$.

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